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TECHNICAL REPORT

Basic Research on Remote Sensing of Fissile Materials utilizing Gamma- rays and Neutrons

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February 2017

HDTRA1-09-1-0059

David C. Ingram

Prepared by:
Ohio University
105 Research and Technology
Center
Athens, OH 45701

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UNIT CONVERSION TABLE

U.S. customary units to and from international units of measurement^{*}

U.S. Customary Units	<div style="display: flex; align-items: center; justify-content: center;"> <div style="margin-right: 10px;"> </div> Multiply by </div> <div style="display: flex; align-items: center; justify-content: center;"> <div style="margin-right: 10px;"> </div> Divide by[†] </div>	International Units
Length/Area/Volume		
inch (in)	2.54 $\times 10^{-2}$	meter (m)
foot (ft)	3.048 $\times 10^{-1}$	meter (m)
yard (yd)	9.144 $\times 10^{-1}$	meter (m)
mile (mi, international)	1.609 344 $\times 10^3$	meter (m)
mile (nmi, nautical, U.S.)	1.852 $\times 10^3$	meter (m)
barn (b)	1 $\times 10^{-28}$	square meter (m ²)
gallon (gal, U.S. liquid)	3.785 412 $\times 10^{-3}$	cubic meter (m ³)
cubic foot (ft ³)	2.831 685 $\times 10^{-2}$	cubic meter (m ³)
Mass/Density		
pound (lb)	4.535 924 $\times 10^{-1}$	kilogram (kg)
unified atomic mass unit (amu)	1.660 539 $\times 10^{-27}$	kilogram (kg)
pound-mass per cubic foot (lb ft ⁻³)	1.601 846 $\times 10^1$	kilogram per cubic meter (kg m ⁻³)
pound-force (lbf avoirdupois)	4.448 222	newton (N)
Energy/Work/Power		
electron volt (eV)	1.602 177 $\times 10^{-19}$	joule (J)
erg	1 $\times 10^{-7}$	joule (J)
kiloton (kt) (TNT equivalent)	4.184 $\times 10^{12}$	joule (J)
British thermal unit (Btu) (thermochemical)	1.054 350 $\times 10^3$	joule (J)
foot-pound-force (ft lbf)	1.355 818	joule (J)
calorie (cal) (thermochemical)	4.184	joule (J)
Pressure		
atmosphere (atm)	1.013 250 $\times 10^5$	pascal (Pa)
pound force per square inch (psi)	6.984 757 $\times 10^3$	pascal (Pa)
Temperature		
degree Fahrenheit (°F)	$[T(^{\circ}\text{F}) - 32]/1.8$	degree Celsius (°C)
degree Fahrenheit (°F)	$[T(^{\circ}\text{F}) + 459.67]/1.8$	kelvin (K)
Radiation		
curie (Ci) [activity of radionuclides]	3.7 $\times 10^{10}$	per second (s ⁻¹) [becquerel (Bq)]
roentgen (R) [air exposure]	2.579 760 $\times 10^{-4}$	coulomb per kilogram (C kg ⁻¹)
rad [absorbed dose]	1 $\times 10^{-2}$	joule per kilogram (J kg ⁻¹) [gray (Gy)]
rem [equivalent and effective dose]	1 $\times 10^{-2}$	joule per kilogram (J kg ⁻¹) [sievert (Sv)]

^{*} Specific details regarding the implementation of SI units may be viewed at <http://www.bipm.org/en/si/>.

[†] Multiply the U.S. customary unit by the factor to get the international unit. Divide the international unit by the factor to get the U.S. customary unit.

Final Report, November 2013
Grant No. HDTRA1-09-1-0059

Title: Basic Research on Remote Sensing of Fissile Materials utilizing Gamma-rays and Neutrons

PI and Institution Address

David C. Ingram
Edwards Accelerator Laboratory
Department of Physics and Astronomy
Ohio University
Athens, OH 45701

1.1 Objectives

- Task 1: Complete measurements of $^{11}\text{B}(\text{d},\text{n}\gamma)^{12}\text{C}$ (Year 1)
- Task 2: Identify possible self-collimated reactions (Years 1,2, 3)
- Task 3: Investigate other possible reactions for measurement (Years 1, 2, 3)
- Task 4: Measure the cross-section of the $^{19}\text{F}(\text{p},\alpha\gamma)^{16}\text{O}$ reaction (Year 2)
- Task 5: Measure the breakup cross-section of the $^2\text{H}(\text{d}, \text{pn})^3\text{He}$ reaction (Year 3)
- Task 6: Measure the cross-sections for additional reactions of interest (Option years 1 and 2, the 4th and 5th years of the proposal)
- Task 7: Annual reports, attendance at conference, annual technical review, and scientific exchange

1.2 Accomplishments

Task 1: Cross-section measurements of $^{11}\text{B}(\text{d},\text{n}\gamma)^{12}\text{C}$

Work began with the measurement of the cross-section of the $^{11}\text{B}(\text{d},\text{n}\gamma)^{12}\text{C}$ reaction. However, we immediately encountered problems with a bismuth germanate gamma-ray detector that we planned to use. We requested, and received, a budget change to allow us to purchase a new detector. Once we received the new bismuth germanate gamma-ray detector, we repeated and extended our measurements of the 15.1 MeV gamma ray yield of the $^{11}\text{B}(\text{d},\text{n}\gamma)^{12}\text{C}$ reaction, as shown in Figure 1. We also established the precision of our measurements. The cross-section for 15.1 MeV gamma emissions from the $^{11}\text{B}(\text{d},\text{n}\gamma)^{12}\text{C}$ reaction is about a factor of two lower than previous (50 year old) measurements^{1,2}.

¹R.W. Kavanagh and C.A. Barnes, "Boron Plus Deuteron Reaction", Physical Review 112 (1958) 503.

²H-M Kuan, P.R. Almond, G.U. Din, T.W. Bonner, "The 15.1 MeV gamma-ray excitation functions of the reactions $^{10}\text{B}(\text{}^3\text{He},\text{p}\gamma 15.1)^{12}\text{C}$, $^{11}\text{B}(\text{d},\text{n}\gamma 15.1)^{12}\text{C}$, and $^{13}\text{C}(\text{}^3\text{He},\alpha\gamma 15.1)^{12}\text{C}$ ", Nuclear Physics 60 (1964) 509.

The detector was heavily shielded with lead to ensure that only gamma rays coming directly from the target were detected. Many gamma rays are produced through interaction of neutrons from the target with the surroundings. The shielding cuts the background down by at least an order of magnitude, which together with use of a pulsed beam and time of flight discrimination enables a minimal background in the 15 MeV region of the spectrum. One other significant improvement we have made, because of the need to cut down the gamma ray count, is in the design of the target stage. We have been able to reduce the mass of the target stage significantly by fabricating the target mount out of 0.5 mm, 99.998% pure aluminum. The machinists in our workshop were able to spin a cup out of this material which become the holder of the boron target. It was pushed over a viton O'ring in on a nylon insulator that housed the secondary electron suppression for the target and the final aperture for the beam. The beam was carefully focused so that as little as 10% of the beam was incident on the final aperture, which was made of tantalum. The effect of all of this was to reduce the gamma ray background significantly, particularly because the neutrons produce by this reaction could not produce as many gamma rays as they interacted with the materials of the target assembly. This enabled data to be taken even with the current detector, with care. As reported in the DTRA review in 2009, we are also able to obtain much better quality data on the 15.1 MeV gamma ray yield through the use of time of flight controlled data acquisition to exclude gamma rays from the spectrum that do not come from the boron target.

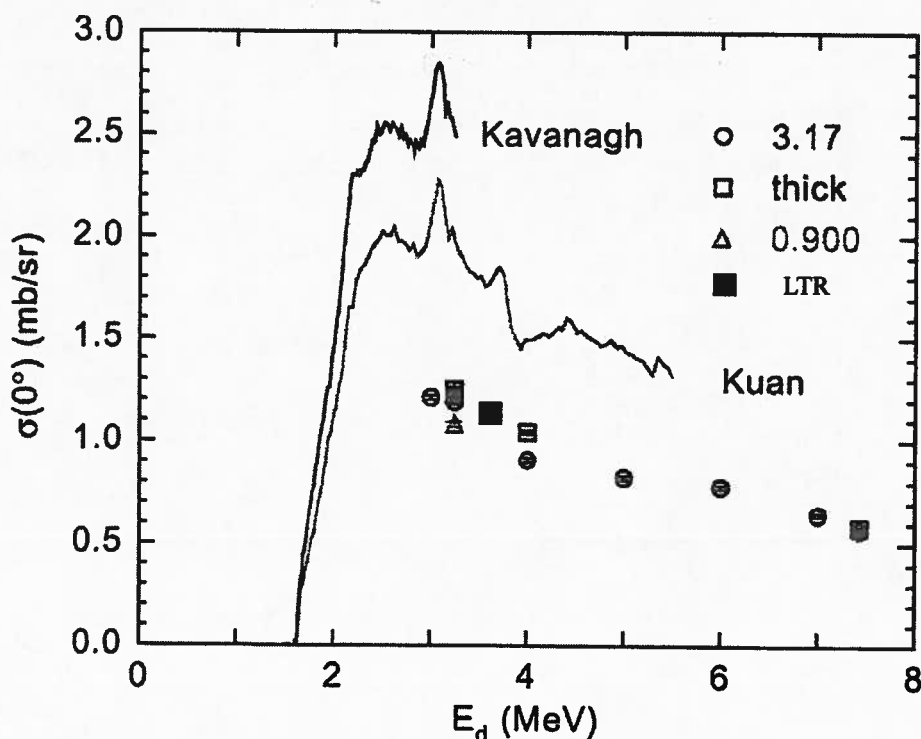


Figure 1 Cross-section for $^{11}\text{B}(d,\gamma n)^{12}\text{C}$ reaction. Lines are from Kavanagh and Barnes, and Kuan et al. The points are from our measurements. The value at 3.5 MeV is 1.18 mb/sr with an uncertainty of $\sim 7\%$.

Task 2: Identify possible self-collimated reactions

In the second year of the program, we were able to find an undergraduate to work on the identification of self collimating reactions. A list of all reactions having a negative Q-value, a half-life greater than 12 years, and an atomic mass number less than 60 was initially created for the proposal. From that list the student eliminated all reactions with an opening energy near or above our maximum beam energy available to us. Edwards Accelerator Laboratory houses a 4.5MeV tandem accelerator which can ideally accelerate beams to energies of $4.5(n + 1)$ MeV where n is the projectile's charge state. The maximum possible charge state varies with the considered nuclei, and it is rare to observe a situation with $n > 3$. In practice the attainable energies are somewhat lower than what could ideally be achieved. All reactions with exceptionally low differences (~ 20 keV) between projectile energy at threshold and at a 45° collimation cone were discarded. From the remaining reactions, an effort was made to locate experimental cross section measurements. Information was often not available on reactions in their originally considered form, but could be found for the kinematically inverted reaction. In these instances an energy conversion was made to allow for use of this data. In the instance that no cross sectional data existed on a reaction in any form, the coulomb barrier was evaluated and compared to the Q-value. This was used to judge whether a given reaction could potentially produce a useable cross section. After all of this data had been compiled and considered, seven reactions remained as promising sources of kinematically-collimated neutrons. These are $^1\text{H}(t,n)^3\text{He}$, $^3\text{H}(^4\text{He},n)^6\text{Li}$, $^1\text{H}(^7\text{Li},n)^7\text{Be}$, $^7\text{Li}(p,n)^7\text{Be}$, $^1\text{H}(^{14}\text{C},n)^{14}\text{N}$, $^4\text{He}(^6\text{Li},n)^9\text{B}$, and $^4\text{He}(^7\text{Li},n)^{10}\text{B}$.

Lithium figures prominently in these reactions and, for some reactions due to the beam intensity limitations on our accelerator for lithium beams, or because the inverse reaction enables access to the reaction within the energy limitations of our accelerator, a lithium target would be advantageous.

The two undergraduates have worked on preparing a lithium target. They researched the literature and decided that a lithium film grown on a nickel substrate and covered with a nickel film would be a good approach. They have learnt how to prepare thin films samples by evaporation for the lithium and magnetron sputtering for the nickel film. They successfully fabricated a target. Investigation of these reactions would have continued under Task 6 if funding had been available..

Task 3: Investigate other possible reactions for measurement

We initiated a collaboration with Dr. Terry Taddeucci of Los Alamos National Laboratory on other nuclear reactions of interest. He performed a series of experiments at Ohio University in the week of August 25th, 2011, to measure the neutron and gamma emissions from the following reactions: $^{13}\text{C}(^3\text{He},n)^{15}\text{O}$, $^{12}\text{C}(^3\text{He},n)^{14}\text{N}$, $^{18}\text{O}(^3\text{He},n)^{20}\text{Ne}$, $^{17}\text{O}(d,n)^{18}\text{F}$, $^{23}\text{Na}(d,n)^{24}\text{Mg}$, $^{11}\text{B}(d,n)^{12}\text{C}$.

A summary of the data has been assembled by Dr. Taddeucci in Los Alamos Report # LA-UR-11-06110. Briefly, the gamma ray yield near 15 MeV relative to that for a ^{11}B target was about a factor of ten lower for $^3\text{He}+^{13}\text{C}$ and a factor of hundred lower for $d+^{14}\text{N}$. The yield from ^{17}O and ^{18}O targets was about a factor of thousand lower. Two figures taken from the report are attached as Appendix A to this report. They show the overlap between the gamma spectra and the photofission cross-section for ^{235}U , and the product of the photofission cross-section and the integral gamma yield. The latter shows that despite the low yield near 15 MeV, $d+^{14}\text{N}$ and

$^3\text{He}+^{13}\text{C}$ are only about a factor of two lower in utility, relative to $\text{d}+^{11}\text{B}$.

Task 4: Measure the cross-section of the $^{19}\text{F}(\text{p},\alpha\gamma)^{16}\text{O}$ reaction

Measurements have been made of the 7.1 MeV gamma emission from this reaction. They have been compared to data obtained by Micklich et al.³ which was also obtained at Ohio University but on a different beam line, with a different gas cell and detector.

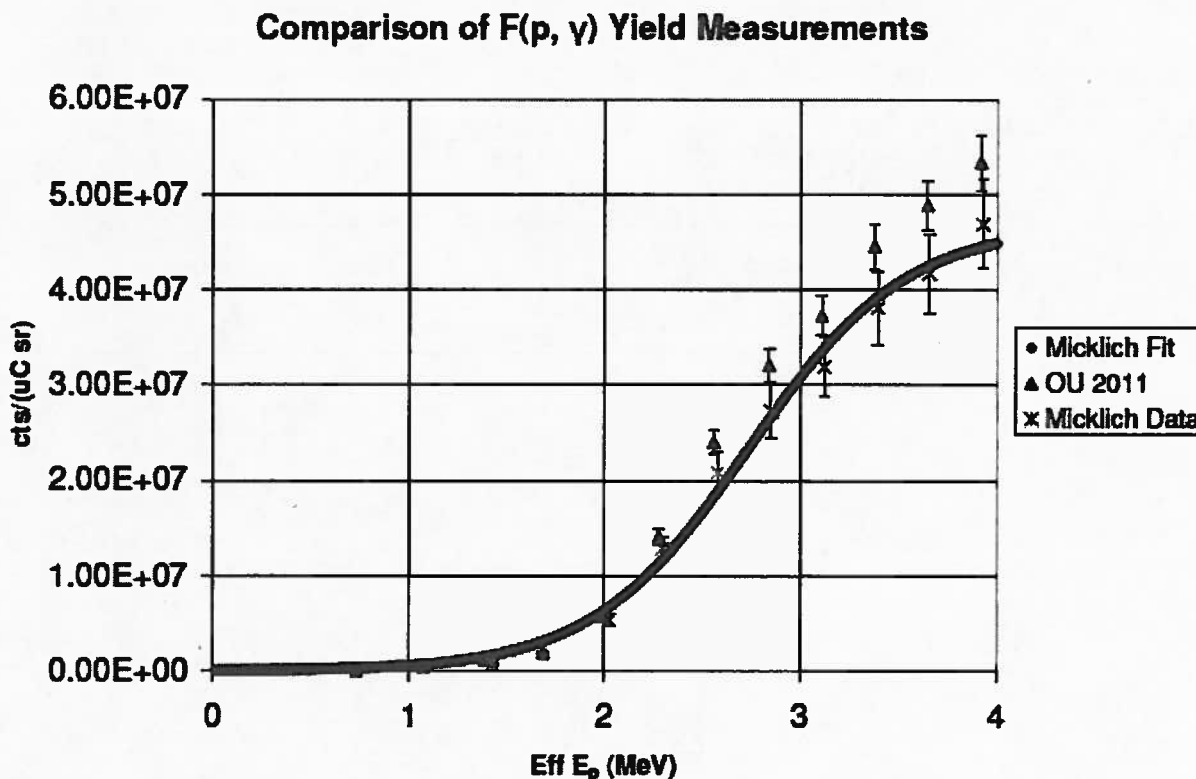


Figure 2 Gamma emission from the F(p,γ) reaction.

There is a difference in the measurement, but this is within the error estimates of the two experiments.

Task 5: Measure the breakup cross-section of the $^2\text{H}(\text{d},\text{n})^3\text{He}$ reaction

In order to measure the breakup cross-section below 1 MeV, lithium-glass neutron detectors were used to measure the neutron production from $^2\text{H}(\text{d},\text{pn})\text{n}$ breakup. The breakup cross-section was measured down to a neutron energy of 250 keV. This was done as a function of emitted neutron angle using a pulsed beam of 6.8 MeV deuterons onto a 3 cm long gas cell filled with deuterium gas.

³ Micklich, B.J., et.al. Measurement of thick-target high-energy γ -ray yields from the $^{19}\text{F}(\text{p},\alpha\gamma)^{16}\text{O}$ reaction. Nuclear Instruments and Methods in Physics Research A. 505 (2003) 1-4.

Shown in Figure 3 are key data taken with the lithium glass detectors and contained in the Master's Thesis of Andrea Richard, the graduate student that took and analyzed the data⁴.

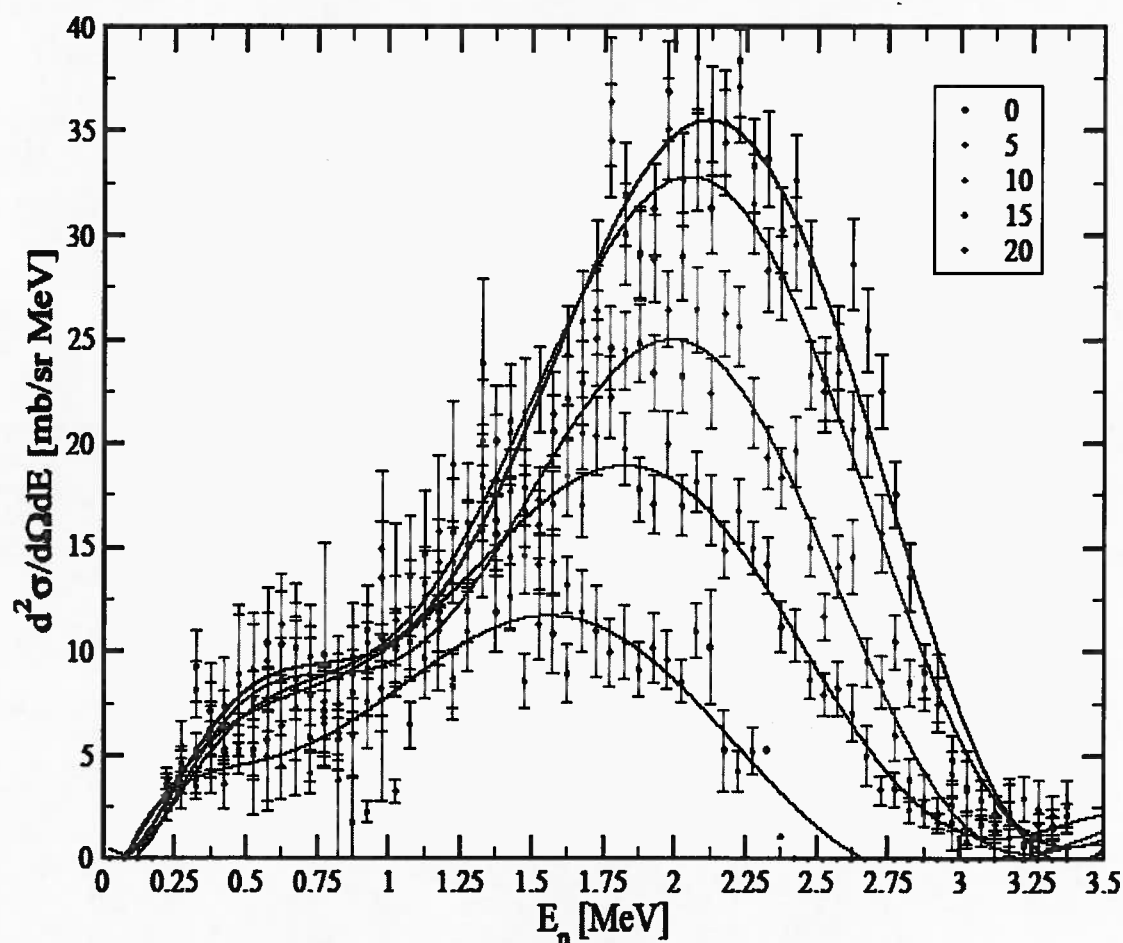


Figure 3 The neutron yield from the $D(d, np)D$ reaction from the lithium glass detector for angles 0 to 20 . Solid lines represent 7th-order polynomial fits which are used to guide the eye.

⁴ Measurement of the Breakup Cross Section of the $D(d, n)$ Reaction at 6.8-MeV for the Active Interrogation of Hidden Fissile Materials, Andrea L. Richard, M.S. Thesis, Ohio University 2013

Shown in figure 4 are the data from the NE213 detectors, There is a significant loss of data below about 1.5 MeV as compared to the lithium glass detector, for that reason no error bars have been included in this figure.

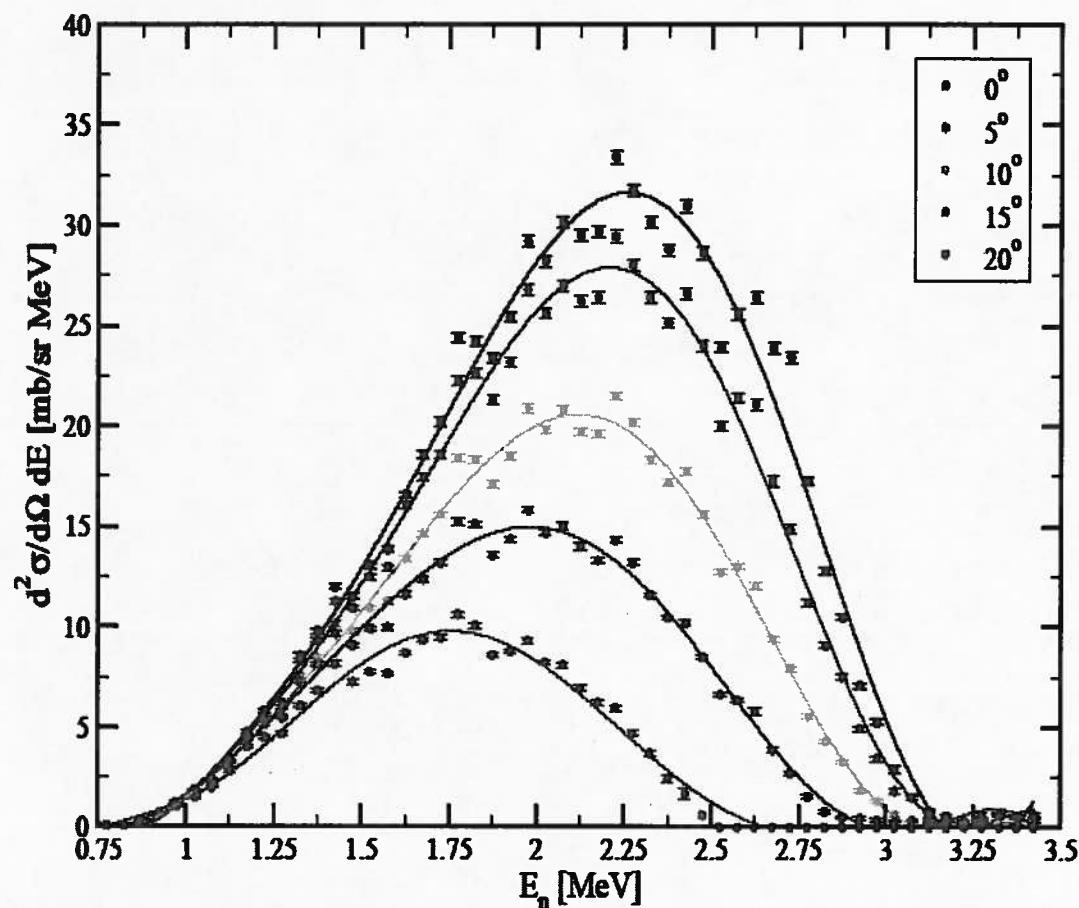


Figure 4 Neutron breakup cross-section data taken with the NE213 detector, compared to Figure 3 there is a significant loss of data below about 1.5 MeV.

Task 6: Measure the cross-sections of additional reactions

From the work on highly collimated reactions we have seven possible reactions, see Task 2 report above. Many of these reactions involve the use of a lithium target. We have worked on the development of such a target. Unfortunately, funding is not available for the option years on this contract so we did not explore these reactions.

Task 7: Annual reports, attendance at conference, annual technical review, and scientific exchange

Annual reports have been submitted and reports of the work have been given at the annual technical review.

1.3 Training opportunities.

Four undergraduates have been supported through summer internships by this grants, one for two summers. They have worked on; neutrons from kinematically collimated reactions, lithium target design and production for kinematically collimated neutron generating reactions, optimizing the ion beam optics. Two graduate students have worked on this program while working on masters projects. Specifically, the measurement of the reactions cross-sections for $^{11}\text{B}(\text{d},\text{n}\gamma)^{12}\text{C}$, $^{19}\text{F}(\text{p},\alpha\gamma)^{16}\text{O}$, and $^2\text{H}(\text{d},\text{n})^3\text{He}$.

Through our collaboration with Terry Taddeucci we were able to expose our students to the work of a National Laboratory. It was very useful for them to meet and work along side such people. Many of our US graduate students have worked as postdocs at National Laboratories as well as obtained permanent positions at these Laboratories.

1.4 Dissemination of Results

In addition to the annual DTRA review in Washington, the graduate and undergraduate students have presented posters of their work at the Ohio University Research and Creative Activities Fair in the Spring, at which they all won awards. As noted elsewhere, two publications have been produced.

2.0 Products

2.1. Publications, conference papers, and presentations:

Measurements of the $^{11}\text{B}(\text{d}, \text{n}\gamma)^{12}\text{C}$ differential cross-section on thick and thin targets, K.W. Cooper, T.N. Massey, D.E. Carter, D.C. Ingram, Nuclear Instruments & Methods in Physics Research Section B-beam Interactions with Materials and Atoms 305 45 2013 DOI: 10.1016/j.nimb.2013.04.048, JUN 15 2013

Comparing modern measurements of the $^{11}\text{B}(\text{d},\text{n}\gamma)^{12}\text{C}$ excitation function with previous values, Kevin W. Cooper, Thomas N. Massey, and David C. Ingram, AIP Conf. Proc. 1525, 709 (2013); doi: 10.1063/1.4802419

References

¹R.W. Kavanagh and C.A. Barnes, "Boron Plus Deuteron Reaction", Physical Review 112 (1958) 503.

²H-M Kuan, P.R. Almond, G.U. Din, T.W. Bonner, "The 15.1 MeV gamma-ray excitation functions of the reactions $^{10}\text{B}(^3\text{He}, p\gamma)^{12}\text{C}$, $^{11}\text{B}(d, n\gamma)^{12}\text{C}$, and $^{13}\text{C}(^3\text{He}, \alpha\gamma)^{12}\text{C}$ ", Nuclear Physics 60 (1964) 509.

³ Micklich, B.J., et.al. Measurement of thick-target high-energy γ -ray yields from the $^{19}\text{F}(p, \alpha\gamma)^{16}\text{O}$ reaction. Nuclear Instruments and Methods in Physics Research A. 505 (2003) 1-4.

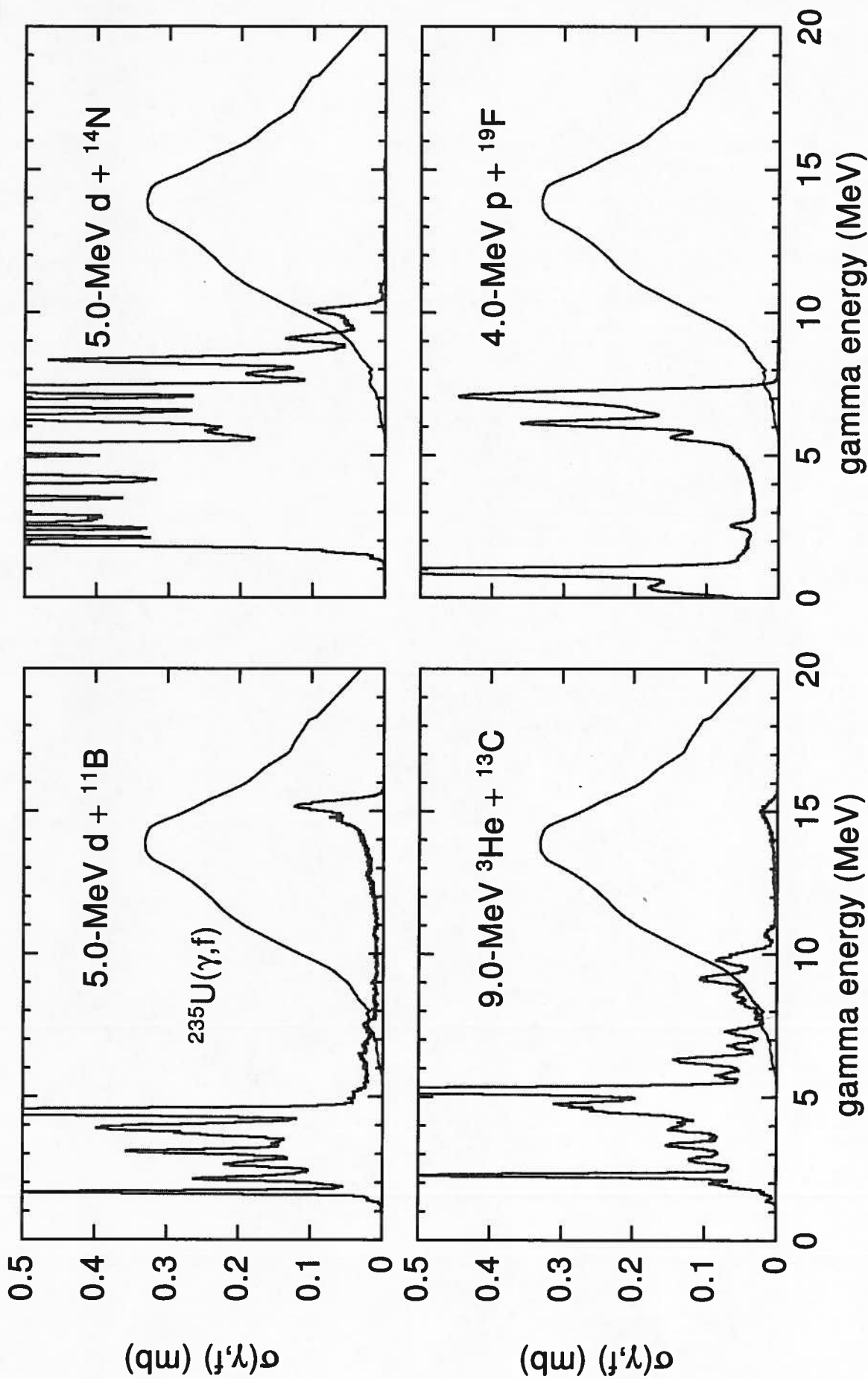
⁴ Measurement of the Breakup Cross Section of the D(d; n) Reaction at 6.8-MeV for the Active Interrogation of Hidden Fissile Materials, Andrea L. Richard, M.S. Thesis, Ohio University 2013

Appendix A

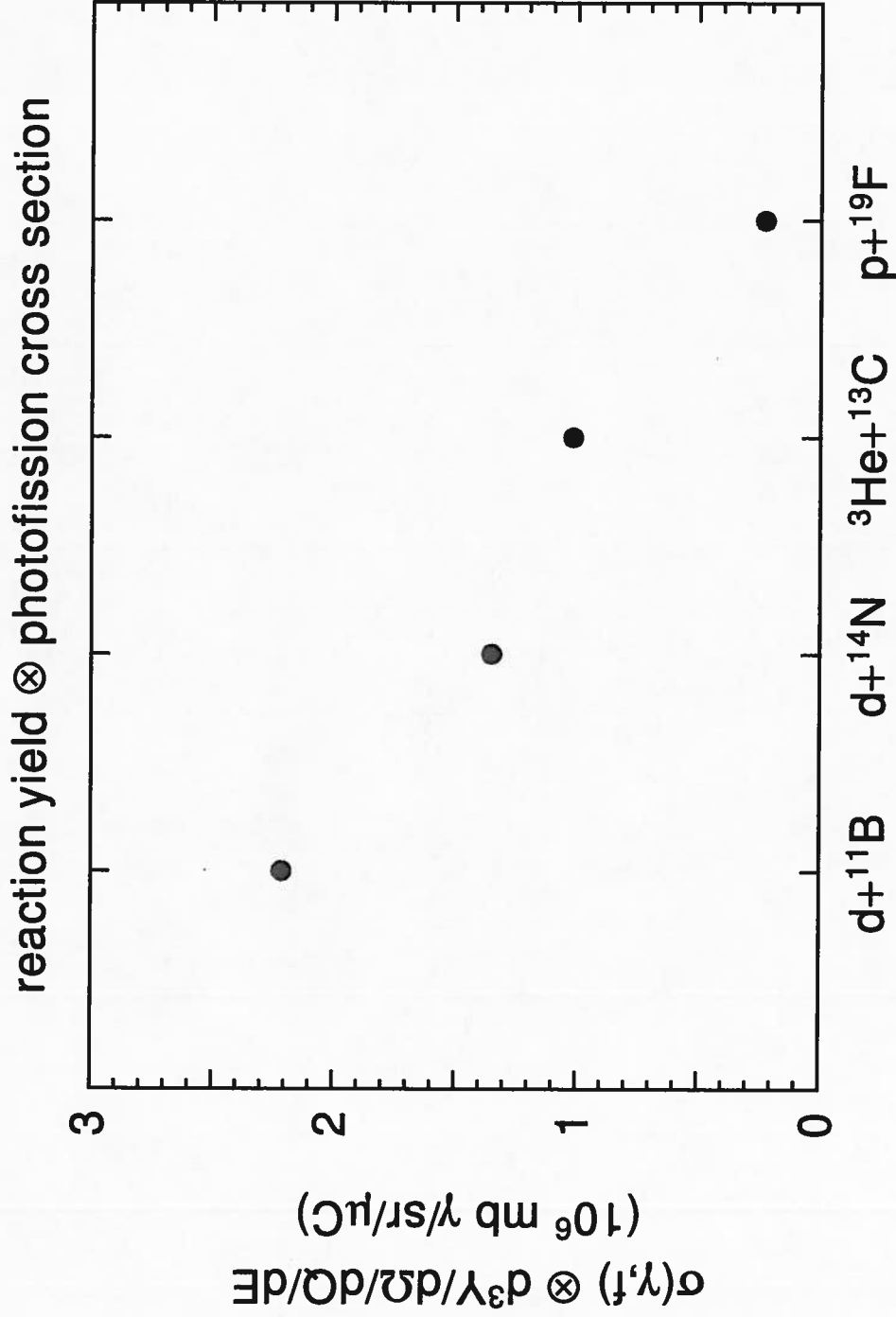
Gamma ray spectra compared to photofission cross-section for ^{235}U .

Taken from "Neutron and gamma-ray output measurements with deuteron and ^3He beams"; T. Taddeucci, B. Perdue, Los Alamos National Laboratory report # LA-UR

Photofission overlap



Photofission cross section \times reaction-yield integral



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